SYNTHESIS AND CHARACTERISATION OF LEAD FREE PIEZOELECTRIC NBT-BT CERAMIC

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By

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Rourkela

CERTIFICATE

This is to certify that the thesis entitled, **"Synthesis and characterization of lead free NBT-BT Ceramic"** submitted by **Mr. Kapil Saxena** in partial fulfillment of the requirements of the award of Bachelor of Technology Degree in Ceramic Engineering at the National Institute of Technology, Rourkela is an authentic work carried out by him under my supervision and guidance.

To the best of my knowledge, the matter embodied in the thesis has not been submitted to any other university / institute for the award of any Degree or Diploma.

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CONTENTS

Abstract

This project dealt with the preparation of Lead-free piezoelectric ceramics (1- x)$Bi_{0.5}Na_{0.5}TiO_3$ - x$BaTiO_3$ that have been prepared by a conventional solid state reaction method , four different composition of (1- x)$Bi_{0.5}Na_{0.5}TiO_3$ -x$BaTiO_3$ were prepared in which x have values 0, 0.04,0.06,0.08. After the addition of $BaTiO_3$ into $Na_{0.5}Bi_{0.5}TiO_3$, the ceramics exhibit strong piezoelectric property at MPB, the ceramics with x = 0.06 exhibit optimum piezoelectric and dielectric properties. X-ray diffraction analysis of the different composition has been carried out to know about the phases. Dielectric measurement was carried out for different compositions. The samples were poled at a voltage 3-4 KV and the piezoelectric measurements were carried out. The micro-structural characterization was done by Scanning Electron Microscopy.

CHAPTER 1

1. INTRODUCTION:

Piezoelectricity is the ability of some materials (crystals and certain ceramics) to generate an electric field or electric potential in response to applied mechanical stress [1]. The effect is closely related to a change of polarization density within the material's volume. If the material is not short-circuited, the applied stress induces a voltage across the material. The piezoelectric effect is reversible in that materials exhibiting the direct piezoelectric effect (the production of an electric potential when stress is applied) also exhibit the reverse piezoelectric effect (the production of stress or strain when an electric field is applied). For example, lead zirconate titanate crystals will exhibit a maximum shape change of about 0.1% of the original dimension.

The piezoelectric effect finds useful applications such as the production and detection of sound, generation of high voltages, electronic frequency generation, microbalances and ultra fine focusing of optical assemblies. Lead-based piezoelectric ceramics with perovskite structure based on lead zirconate titanate (PZT) are widely used for actuators, sensors as well as microelectronic devices because of their excellent piezoelectric properties. However, because of the high toxicity of lead oxide, the use of the lead-based ceramics has caused serious lead pollution and environmental problems. Therefore, it is necessary to develop lead-free piezoelectric ceramics for replacing them.

1.1 Sodium Bismuth Titanate:

Sodium bismuth titanate is a dielectric material which is being used widely due to its high temperature dielectric constant and it is a lead free material. $Na_{0.5}Bi_{0.5}TiO_3$ (NBT) is a ferroelectric complex having Bi^{3+} and Na^{+} on the A-site of ABO_3 perovskite structure with a rhombohedral symmetry [2]. It is considered as one of the good candidates for lead-free piezoelectric ceramics because of a large remnant polarization at room temperature [3].

However, it also has a high coercive field making the poling of the ceramic difficult. Thus the NBT ceramic usually exhibits weak piezoelectric properties, high dielectric loss and difficult to polarize [4]. To improve the piezoelectric properties doping of some materials are used and a number of NBT-based solid solutions, such as NBT- $Bi_{0.5}K_{0.5}TiO_3$ [5, 6], NBT-$NaNbO_3$ [7], Bi_2O_3- doped NBT [8], $Bi_{0.5}(Na_{1-x-y}\ K_xLi_y)_{0.5}TiO_3$ [9-12], have been developed and studied intensively. It is also noted that as a classical NBT-based system, the piezoelectric properties of NBT-$BaTiO_3$ (NBT-BT) ceramics is frequently reported [13-15]. However, there is little report on the depolarization temperature T_d and temperature dependences of electrical properties for NBT-BT ceramics. In the present work, (1-x) $Bi_{0.5}Na_{0.5}TiO_3$ -x$BaTiO_3$ lead-free ceramics were fabricated by conventional solid state synthesis method and their structure, densification, electrical properties were studied systematically.

1.3 Structure of Sodium Bismuth Titanate:

Bismuth sodium titanate is an ABO_3 distorted perovskite with an rhombohedral R3c crystal structure at room temperature [16]. The standard ABO_3 perovskite formula for NBT is ($Bi_{0.5}$ $Na_{0.5}$) TiO_3. An ABO_3 perovskite can be considered in two ways; one way is to have the bismuth and sodium cation occupy the corners of a cubic unit cell, oxygen cations occupying the face centers, and a titanium cation in the center of the oxygen octahedra that is formed.

The other way, is a three- dimensional cubic network of 8 corner-sharing TiO_6 octahedra with bismuth and sodium cations at the center of the cube formed by the octahedral [17].

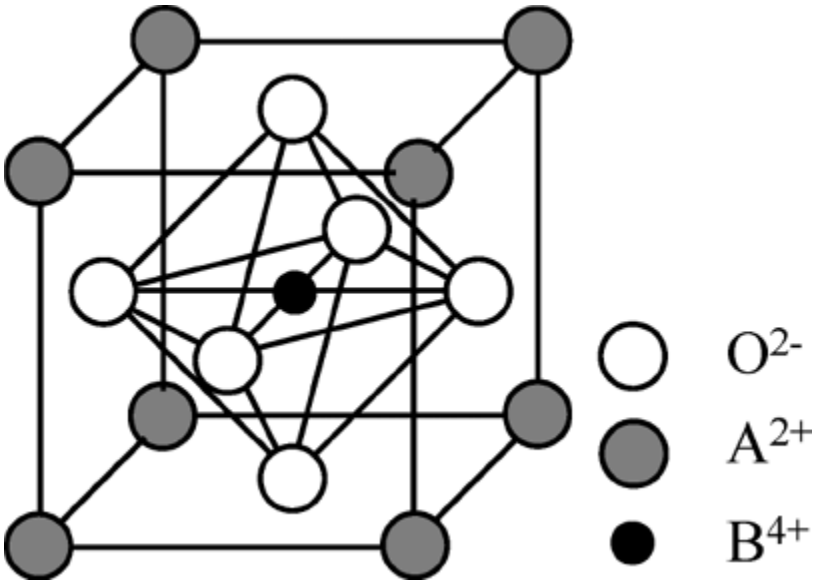


Figure 1: ABO_3 perovskite structure of cubic NBT

Figure 1 represents a typical ABO_3 perovskite, shown here as cubic NBT. The figure suggests that the bismuth and sodium ions are ordered on the A site of the structure; this is only to show the stoichiometry that is present in an ideal mixture. The real material does not exhibit any long range ordering as described later in the text.

1.2 Doping in Sodium Bismuth Titanate:

Many studies have been performed on NBT to determine how dopants affect the structural and electrical properties of the material. Some studies focus on dielectric properties, while others focus on piezoelectric properties. Both A-site and B-site dopants have been studied to determine how they affect the properties of NBT, some of these dopants include (Ba, Pb, Sr, Zr, La, K, Bi) [18-22].

Some of the main drawbacks of this material are that it has a high coercive field and high conductivity. Different dopants can be added to NBT to combat some of its drawbacks, such as to decrease coercive field or increase the piezoelectric constant. The effects of barium on the properties of NBT have been characterized by various groups over the years. Many studies that involve barium doping also include another A or B-site cation to evaluate the effects of multiple-site doping on the properties [23-26]. One of the advantages of doping with barium is that there is a morphotropic phase boundary (MPB) between the rhombohedral and tetragonal phases of the structure. Dielectric materials near an MPB are interesting because they exhibit anomalously large dielectric constant values compared to other compositions.

Most of the papers cited in literature have shown that the system presents a MPB around 6 mol% BT, In the trigonal region, the dielectric behavior of the NBT–BT solid solutions is, as expected, very similar to the one observed for pure NBT or low-lead titanate.

CHAPTER 2

2. Literature Review:

The NBT-BT material system, like several other lead-free materials, was first reported in the 1960s by Smolenskii *et al.* but did not receive much attention until the recent surge in lead-free material development in the past two decades [27]. Some of the initial dielectric and optical property measurements of NBT were reported in the 1990s by various sources [28-30]. Preliminary structural studies of NBT did not provide a definitive structural understanding, but in 2002 Jones and Thomas found that it expresses the rhombohedral *R3c* space group at room temperature and changes to tetragonal and subsequently cubic during heating. NBT is a promising material due to its high Curie temperature and a piezoelectric constant similar to that of BT. But BT has very low curie temperature without addition of lead. Lead additions increase the Curie temperature up to about 150° C; however, lead also de-stabilizes the low temperature phase transitions.

The commonly used materials are lead based eg-lead zirconate titanate (PZT), lead magnesium niobate (PMN), etc. However, the toxicity of lead oxide and its high vapor pressure during the sintering process results in serious environment problems. As a consequence, it becomes necessary to develop low-lead or lead-free piezoelectric materials with properties close to those of the PZT system. Because of environmental issues, new lead-free piezoelectric materials are the object of many studies. The $Na_{0.5}Bi_{0.5}TiO_3$compound (NBT) is considered as a possible candidate for such applications. NBT has large polarization, high temperature dielectric constant and also no lead so it can be developed to be used as future replacement for all lead based compounds. But pure NBT has a drawback of high coercive field and the high conductivity causes problems during the poling process.

To improve the properties some doping has to be done such as NBT–$BaTiO_3$, NBT–$PbTiO_3$, NBT-$K_{0.5}B_{0.5}TiO_3$, NBT–$SrTiO_3$ and NBT–$BiFeO_3$ [31-32].

Elisa Mercadelli et al, proposed that Ba-modified bismuth sodium titanate with composition $0.94[(Bi_{0.5}Na_{0.5})TiO_3]$-$0.06BaTiO_3$ (BNBT) was prepared by a citrate nitrate sol–gel combustion method. The sol was obtained using barium acetate, bismuth nitrate, sodium nitrate and a peroxo-citrate complex of titanium isopropoxide as starting precursors.

Various molar ratios of citrate/nitrate (C/N) were considered for the sol production. The corresponding gels were fired at different temperatures (300, 400, 500 ^{0}C) in order to evaluate the conditions necessary to obtain the decomposition of the precursors and the formation of the pure NBT-BT perovskitic phase in a single step.

The best conditions to obtain the desired phase are: (C/N) = 0.2, and combustion temperature of 500 ^{0}C. The electrical properties are comparable to those reported for conventionally prepared materials [33]. d_{33} for NBT-BT was found to be 125pc/N & Kp was found to be 0.272 and relative permittivity was found Er = 698.

Junjie Hao et al, proposed a stearic acid gel method for the preparation of nanocrystalline single phase NBT powder at relatively low treatment-temperature. It shows that pure single phase NBT powders could be obtained at 700 °C for 1 h, and the particle size is about 20 nm. With an increase in the calcination temperature, crystallite size increased [34].

Comparing with solid-state reaction NBT powders, the structure of NBT nanocrystalline belongs to pure pervoskite type, and no other intermediate phase is found. All NBT particles were very small, with an average size of 10–40 m. All particle size is much smaller than that by the solid-state method. The bulk density of the sintered samples was measured by the Archimedes method. The measured density ratio is 97%, higher than that 90% of the solid phase reaction samples.

Compared with the sample prepared by traditional process, this sample derived from nano NBT is less porous and has the homogeneous microstructure with the grain size of about 200 nm. Piezoelectric and dielectric properties of samples prepared by stearic acid gel was found ε (at 1 MHz) = 446 and d_{33} (PC N^{-1}) = 60.

Nagata et al. reported on the properties of NBT with the addition of both $BaTiO_3$ and $(Bi_{0.5}K_{0.5})TiO_3$ (BKT). They reported that an MPB exists at a composition containing 85.2% NBT, 2.8% $BaTiO_3$ and 12% $(Bi_{0.5}K_{0.5})$ TiO_3. They found that the most useful compositions were located near the MPB because they exhibited the anomalous electrical behavior associated with MPB compositions [35].

Lanfang Gao et al [36] fabricted lead-free $(1-x)BaTiO_3–xBi_{0.5}Na_{0.5}TiO_3$ (x = 0.01, 0.02, 0.05, 0.1, 0.2, 0.3) ferroelectric ceramics by the conventional solid state reaction technique. Sintering was made at 1200 ^{O}C for 2–4 h in air atmosphere.

The dielectric and ferroelectric properties were studied. Room temperature permittivity was found to decrease as $Na_{0.5}Bi_{0.5}TiO_3$ (NBT) content increases. Only the sample with 0.3 mol NBT was found to have relaxer behavior. The Tc shifted slightly only for NBT addition lower than 0.1 mol. The highest Tc (about 150 ^{O}C) was obtained for 0.2 mol NBT addition. The remnant (Pr) decreases whereas the coercive field, Ec, increases monotonously as the NBT content increases.

Man-Soon Yoon et al [37] used pre-synthesized $BaTiO_3$ and pre-milled Bi_2O_3, Na_2CO_3, $BaCO_3$ powders and calcination powder milled with a high energy milling machine in order to obtain a nano-particle size. The second one is a conventional one to compare with the former process. The dielectric and the piezoelectric properties of sintered specimens fabricated by the two different processes were evaluated. It was found that the properties of the nano-sized NBT-BT ceramic increased by the modified mixing and milling method, showing superior characteristics in terms of the piezoelectric, dielectric constant and sintering density compared with those of the conventional process. it was found that the remnant polarization Pr for the nano-sized NBT-BT specimen has a higher value of 37.8 mC/cm2 compared with that of 12 mC/cm2 for conventional NBT-BT; whereas the coercive field (Ec) has a similar value.

Summary of Literature Review

(1) There are different routes as discussed in the literature survey for synthesis of phase pure barium doped sodium bismuth titanate (NBT-BT).

(2) Solid state synthesis route is very easy and cheapest route and effective for preparation of phase of NBT-BT.

(3) But it is very difficult to get sintered density above 94% of theoretical density by conventional solid state route.

(4) By increasing the barium titanate concentration in NBT, piezoelectric and dielectric as well as density of the microstructure increases with BT content.

(5) The dielectric and piezoelectric properties of NBT-BT prepared by conventional solid state route is not much better than compare with other methods.

(6) Conventional solid-state method for synthesizing NBT ceramic powder often format large grains which are difficult to disperse and affect the sintering properties of NBT.

CHAPTER 3

3. Statement of Problem:

1. Develop solid solution of NBT-BT by solid state reaction synthesis route.
2. To observe the variation of density by changing BT dopant concentration on powder prepared by solid state synthesis route
3. XRD analysis for phase identification
4. Sintering of powder compact (pallets)
5. Density measurement and SEM analysis
6. Dielectric and piezoelectric properties measurements

Experimental Procedure

3.1 The raw materials used for synthesis of NBT:

- Bismuth oxide (Bi_2O_3)
- Sodium carbonate (Na_2CO_3)
- Barium carbonate ($BaCO_3$)
- Titanium dioxide (TiO_2)

3.2 Powder preparation by solid state reaction synthesis:

Solid state synthesis method was adopted to produce powder for samples. (1-x)NBT-xBT was prepared with x=0,0.04,0.06,0.08. For powder preparation bismuth oxide (Bi_2O_3) powder, sodium carbonate (Na_2CO_3) powder, titanium dioxide (TiO_2) powder, barium carbonate ($BaCO_3$) was used. The powders were weighed and mixed properly in a mortar. After mixing in dry condition iso propyl alcohol was added and mixed and again grinding was done till it became dry.

3.3 Calcination of powder:

The powder was grinded and dried properly and then Calcined in alumina crucible at 800^oC for 2 hours. The calcinations help in driving out all volatile and gaseous material from powder.

3.4 X-RAY Analysis of Calcined powder:

Calcined powder of all composition were subjected to phase analysis by X-ray diffraction

(Pan analytical, Phillips, Netherland). This is done to know the different phases present in the Calcined powder. The angle range was 15^{o}-70^{o}.

3.5 Compaction into pellets:

The Calcined powder was mixed with 3% PVA solution (for binding). It was mixed in an agate mortar and left to dry. After drying it was scraped and grounded to fine powder. The different compositions powder were separately packed after being weighed (around 0.7 gms).The powder was then pressed into pellets by uniaxial compaction with load of 4 ton.

3.6 Sintering of pellets:

The compacted pellets were sintered in conventional furnace at 1150^{o}C for 2 hours.

3.7 Measurement of density by Archimedes principle:

The densities of the sintered pellets were measured by Archimedes principle using kerosene oil. The dry weight, soak weight and suspended weight were measured. The densities of pallets were calculated by formula:

Density = {dry weight/ (soak weight – suspended weight)} *0.81

3.8 Micro structural analysis by SEM:

The sintered pellets were taken for SEM analysis. JEOL JSM-6480 SEM instrument was used. The pellets were sputtered in a sputtering unit. Then they were loaded for analysis. This analysis helps us to know the complete microstructure of the sintered sample. Before SEM platinum coating of 20 mÅ was done for 3 minutes by JEOL JFC- 1600 Auto fine coater.

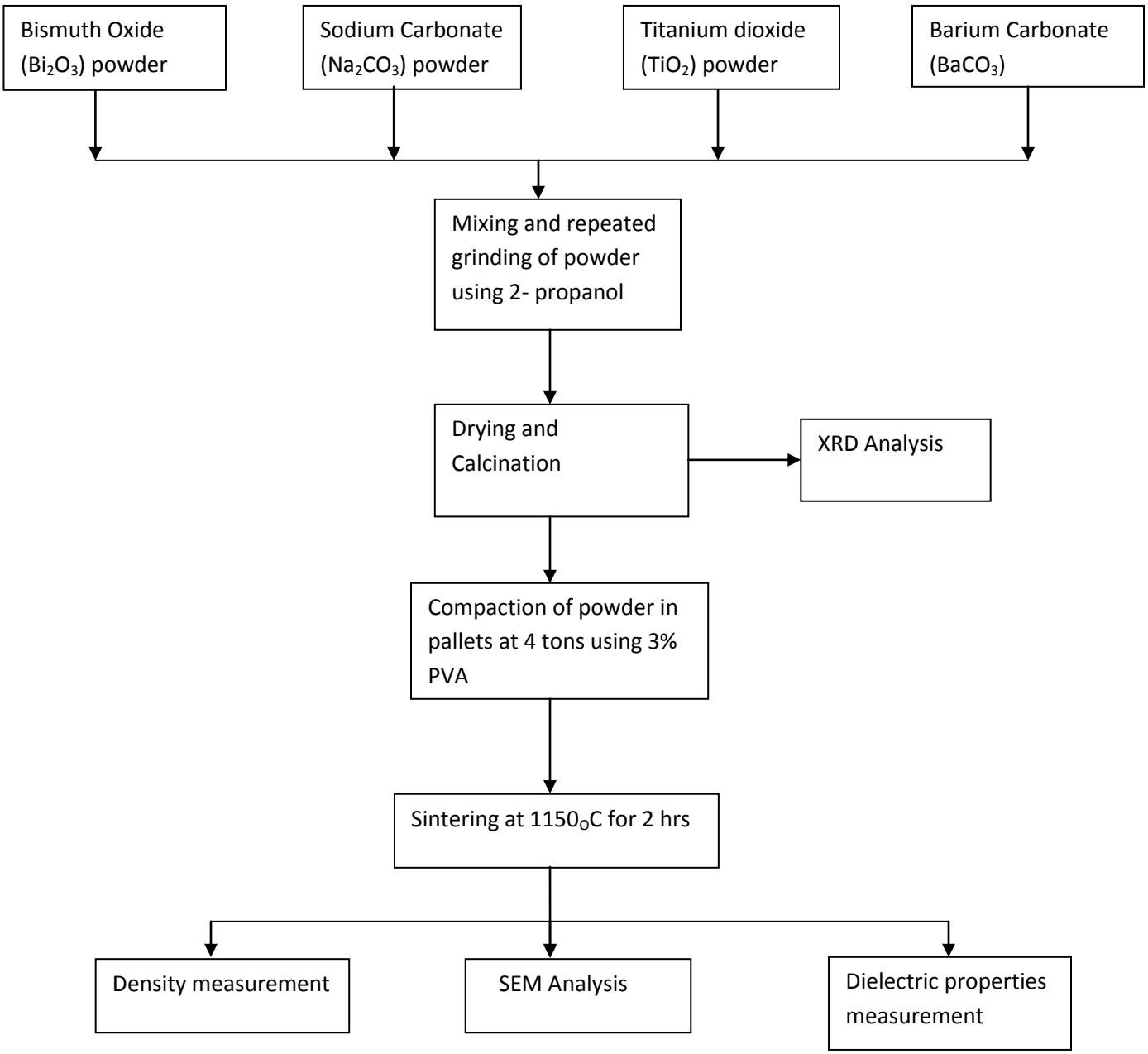
3.9 Preparation of sample for dielectric properties measurement:

Pallets were fine polished by using emery paper 400 & 600 microns and make equal width of pallet over surface using digital calipers. After polishing ultrasonic vibration was given to the samples for 3 minutes by immersing sample in acetone, this process was used for removing air bubbles inside the pallets. Pallets were dried at $80^{O}C$ for 30 minutes.

Silver coating of samples was done by using silver paste and thinner liquid. And samples were again dried for around 80-$90^{O}C$ for 2 hours. Then samples were fired at $500^{O}C$ for 15 minutes. Then dielectric properties was measured by HIOKI 3532-50 LCR Hi tester using frequency 100Hz - 1MHz and d_{33} value was measured by using YE2730A d_{33} meter and poling is done by ntpl series-DHVMN2 instrument.

Before measurement of the piezoelectric constant d_{33} poling of the sample was done because the domains of dipoles are randomly oriented shows no net dipole.

Flow chart of the procedure for preparation of NBT



Preparation of NBT- BT samples: (Example)

1. For 5 gms NBT sample compositions -

Bi_2O_3 = 2.74 gms

Na_2CO_3 = 0.62 gms

TiO_2 = 1.89 gms

2. For 5 gms 0.96 NBT- 0.04 BT:

Na_2CO_3 = 0.595 gms

Bi_2O_3 = 2.63 gms

$BaCO_3$ = 0.169 gms

TiO_2 = 1.883 gms

3. For 5 gms of 0.94NBT – 0.06BT:

Na_2CO_3 = 0.5828 gms

Bi_2O_3 = 2.5756 gms

$BaCO_3$ = 0.2538 gms

TiO_2 = 1.8798 gms

4. For 5 gm s of 0.92NBT – 0.08 BT:

Na_2CO_3 = 0.5704 gms

Bi_2O_3 = 2.5208 gms

$BaCO_3$ = 0.3384 gms

TiO_2 = 1.8764 gms

By adding these compounds and mixing them with propanol-2 we got the desired composition by following above procedure.

CHAPTER 4

4. Results and Discussions:

4.1 XRD Analysis:

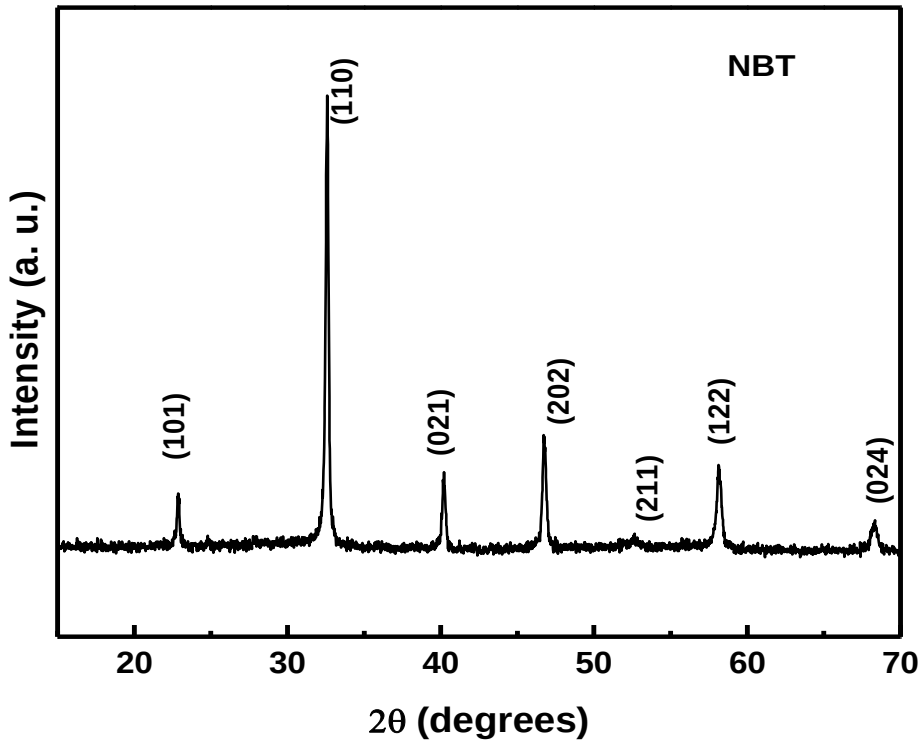


Figure (2) shows XRD pattern of pure NBT Calcined at 800^{O}C

Figure (2) shows the XRD pattern of pure NBT calcined at 800^{O}C for 2h. It was found that there is no impurity peak and match with JCPDS card no 36-0153. The crystal structure was found rhombohedral and the average crystallite size is 43.921 nm.

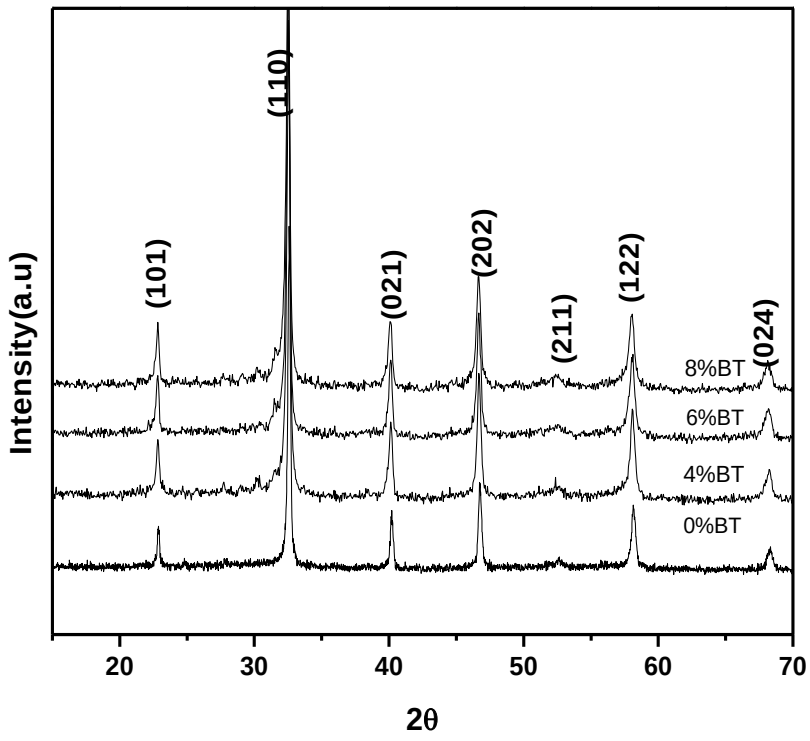


Fig.3 XRD patterns of (1-x) ($Na_{0.5}Bi_{.0.5}TiO3$)-x($BaTiO_3$) prepared by solid state synthesis route calcined at 800^{o}C for 2h

Fig.3 shows the XRD patterns of (1-x) ($Na_{0.5}Bi_{.0.5}TiO3$)-x($BaTiO_3$) prepared by solid state synthesis route calcined at 800^{o}C for 2h . The XRD Pattern shows that sample contains no impurity phase or secondary phase was found .It is matched by JCPDS card no 36-0340. For higher $BaTiO_3$ content there were no impurity phase and the peaks were also get broaden. The average crystallite size was 35-43.9 nm. It was observed that crystallite size was decreasing with addition of barium titanate .

Composition	Crystallite Size (nm)
NBT	43.92
NBT –BT(4%)	43.9
NBT- BT(6%)	35.14
NBT- BT(8%)	35.04

4.2 Bulk density of sintered pellets:

Density was measured by Archimedes principle.

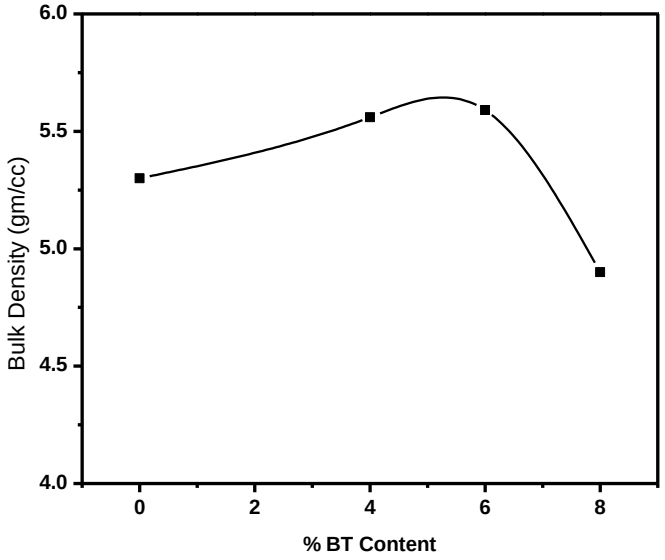


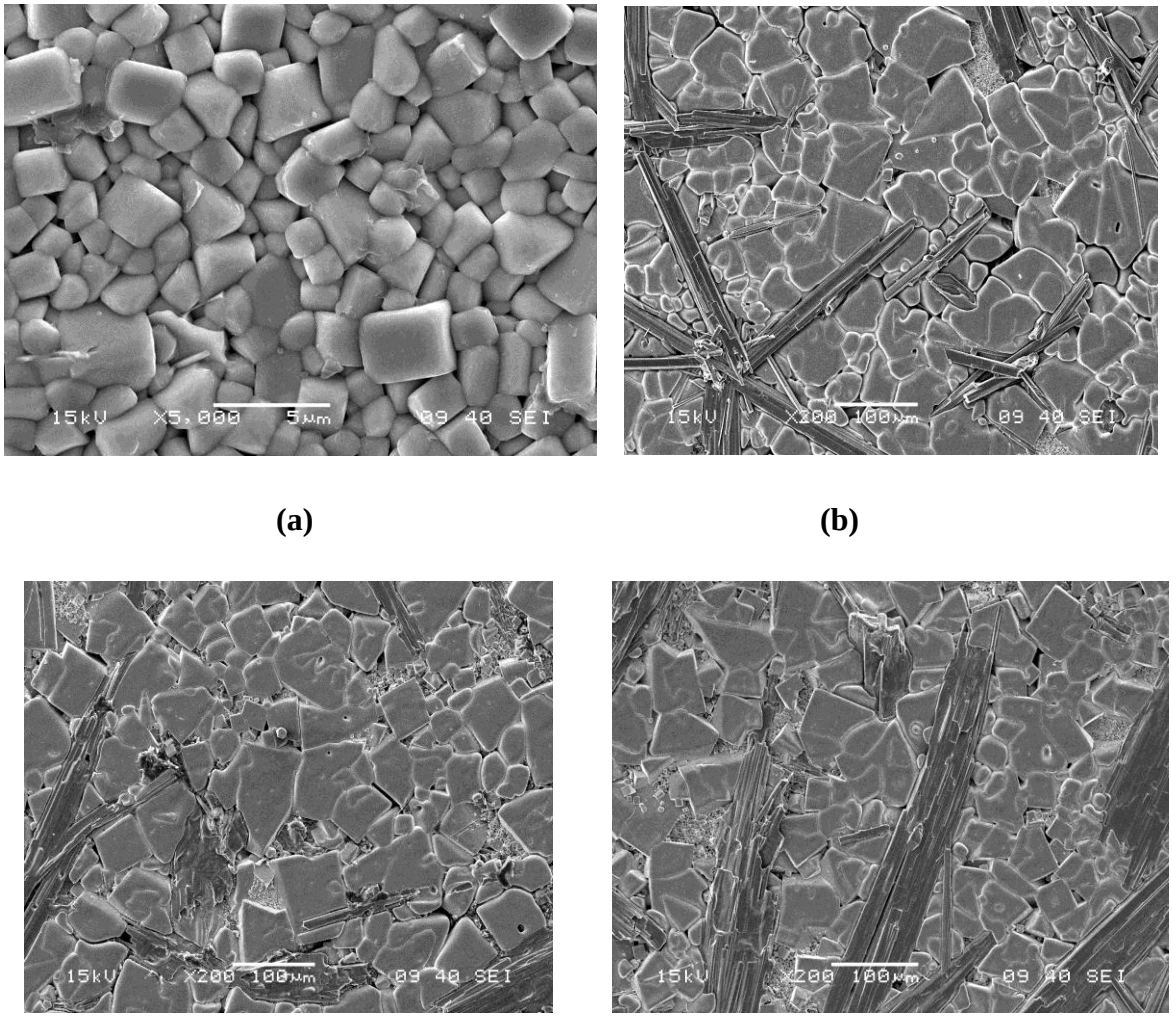
Figure4 Variation of bulk density of sintered sample with BT content

It was found that density of NBT increases with increasing BT content upto 6% and then decreases with further addition. Average density of 0.94NBT- 0.06BT = 5.6 gm/cc. Theoretical density is 5.9 gm/cc, so density percentage = 94.9. Density of NBT-BT was found maximum at 6% BT concentration, sintered at $1150^{O}C$.

4.3 Scanning Electron Microscope Analysis of NBT-BT samples:

4.3(a) SEM of sintered pallets of NBT pure phase:

The figure shows micrograph of NBT-BT solid solution with different BT addition.



(a) **(b)**

(c) ***(d)***

Figure5: shows SEM of (a) NBT (b) 0.96NBT- 0.04BT (c) 0.096NBT-0.06BT (d) 0.092NBT-0.08BT sintered at 1150^{O}C

Fig. 5(a),(b),(c),(d) shows the SEM image of sintered sample of NBT and solid solution of NBT with BT. It is clear from the micrograph that BT addition significantly increases the grain size of the sample. Pure NBT has grain size of 2-5 µm. NBT-BT solid solution has grain size 50-150 µm. Microstructure shows highly dense structure for NBT and NBT-BT solid solution. NBT-BT solid solution contains few plates like grain. Further studies are required to know the composition of the plate like morphology.

4.4 Measurements of the piezoelectric & dielectric properties:

Dielectric properties (dielectric loss, permittivity) of the NBT-BT sample were measured by HIOKI 3532-50 LCR Hi tester and d_{33} value was measured by YE2730A d_{33} meter.

Piezo electric constant d_{33} measurement:

Before measuring d_{33} value poling of samples was done because dipoles in the materials are randomly oriented when electric field is applied for some time then orientation of the domains increase in the direction of electric field. Sample was put in poling machine ntpl for 20-30 minutes then after that d_{33} value was measured.

For pure NBT phase:

Leakage current = 0

Thickness of the sample (pallet) = 1.13 mm

Voltage applied = 3.3 kv for 20 minutes

d_{33} value = 58 pc/N

For 0.96NBT-0.04BT:

Leakage current = 0

Thickness of the sample (pallet) = 1.27 mm

Voltage applied = 4 kv

d_{33} value = 82 pc/N

For 0.94NBT-0.06BT:

Thickness of the sample (pallet) = 1.18 mm

Voltage = 4 kv applied for 20 minutes

d_{33} value = 89 pc/N

For 0.94NBT-0.08BT:

Thickness of the sample (pallet) = 1.35 mm

Voltage applied = 3.6 kv applied for 20 minutes

d_{33} value = 62 pc/N

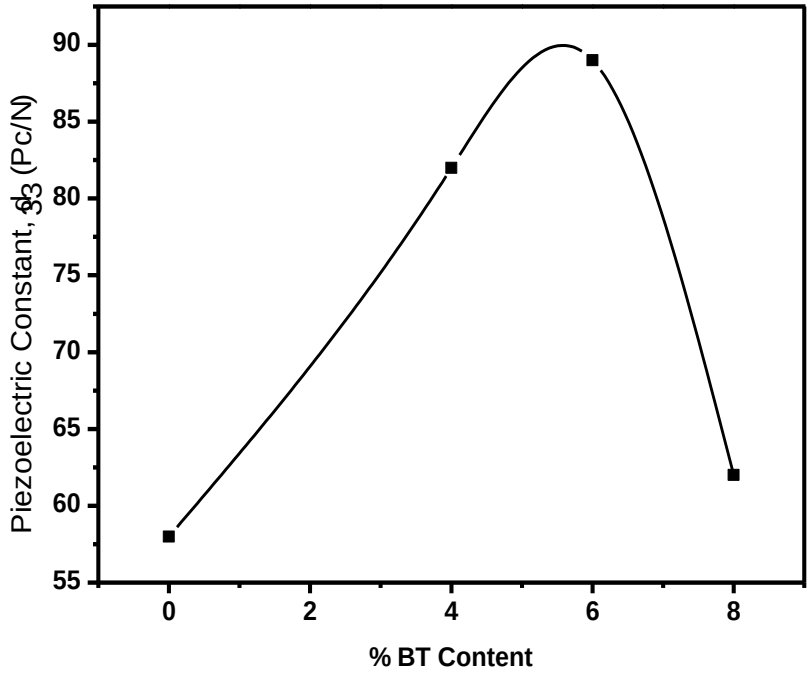


Fig.6 Variation of Piezoelectric constant with %BT addition

The d_{33} increased with increasing BT ($BaTiO_3$) content and decreased after certain limit of BT content. It was found maximum at 6% BT concentration. Enhancement of the d_{33} value can be explained from the formation MPB at 6 mol% BT addition [33, 34].

4.5 Dielectric property of NBT and solid solution:

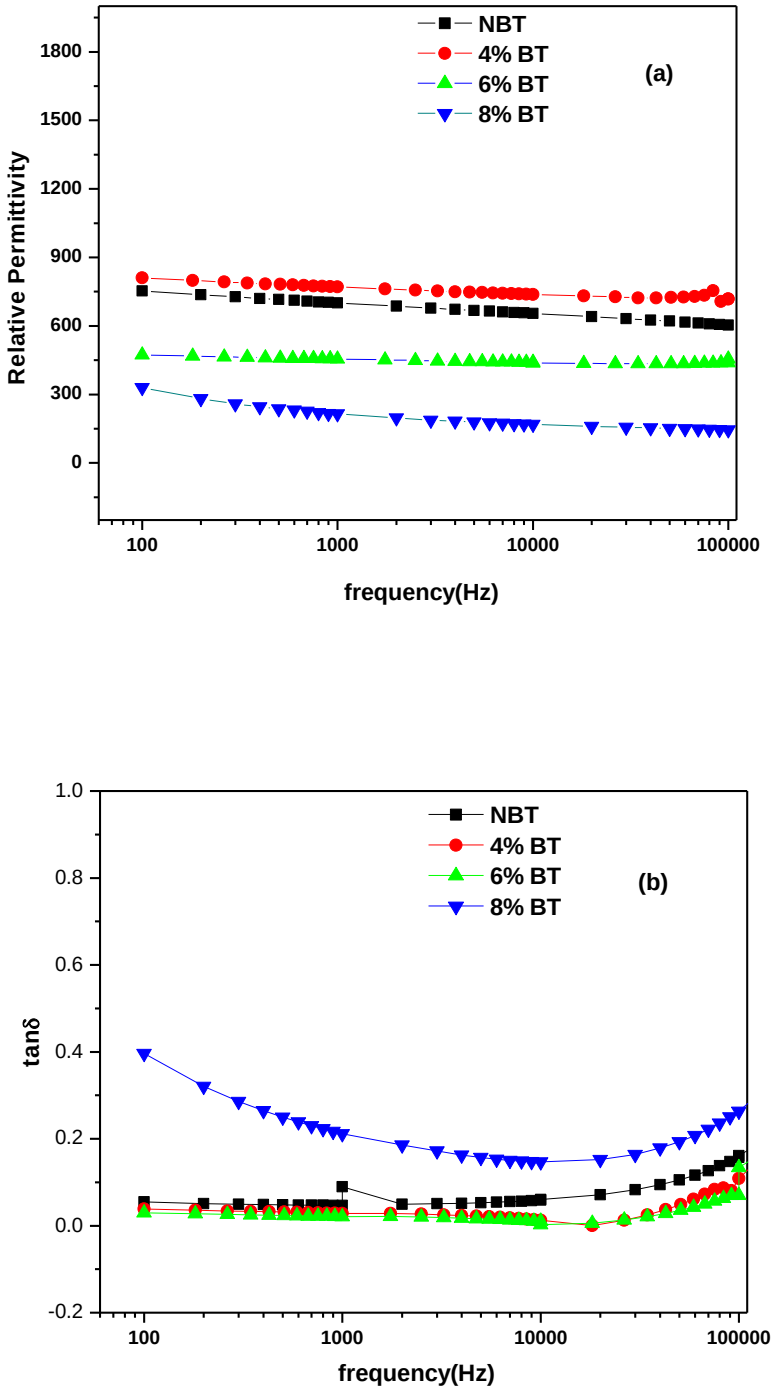


Fig.7 Variation of (a) Relative permittivity (ε) (b) dissipation factor with frequency for NBT-BT ceramics with different BT addition.

The relative permittivity of solid solution increases for 4 mol% BT addition and then decreases with higher BT addition. Above figure shows that the dielectric loss of the solid solution initially decreases with BT addition and again increases for 8 mol% BT addition.

Different properties are summarized in Table 1. It can be seen that the ε and d_{33} values of the specimen of conventional solid state method are less than that of obtained from sol gel and auto combustion method.

Table 1:

Sample Composition	**Density(g/cc)**	**Permittivity ($\acute{\varepsilon}_r$) at 10 kHz**	**Tanδ (loss) at 10 kHz**	**d_{33} (PC/N)**
NBT	5.46	654	0.0589	58
0.96NBT 0.04BT	5.58	738	0.0128	82
0.94NBT 0.06BT	5.6	441	0.0106	89
0.92NBT 0.08BT	4.94	168.8	0.147	62

5. CONCLUSION:

(1-x)$Na_{0.5}Bi_{0.5}TiO_3$-x$BaTiO_3$ ceramics (where x=0, 0.04, 0.06, 0.08) have been prepared by conventional solid state synthesis method. Phase pure ($Na_{0.5}Bi_{0.5}$)TiO_3 powder can be prepared at calcination temperature 800^{o}C. XRD analysis of different composition of NBT-BT has shown that there were no impurity phases.

By density measurement it was observed that density of NBT-BT increases with increasing BT content and was found maximum at 6% BT content. By SEM analysis it was observed that grain size of NBT increases with addition of barium titanate.

Piezoelectric properties(d_{33}) of NBT increased with increasing barium titanate content up to 6% BT then decreases and maximum d_{33} value at 6% barium titanate(d_{33}=89 Pc/ N). Barium titanate also reduced the dielectric loss factor. Dielectric loss (Tan δ) decreases with increasing BT concentration.

6. Future work

- To study the dielectric properties with varying temperature
- To details analysis of the microstructure of solid solution sample

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